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Chapter 4

Atmospheric Science

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4.1 Introduction

The atmospheric science subgroup considered a number of possible experiments which might be carried out under low gravity conditions. These experiments were considered in the light of two basic questions: (1) Why carry out these particular experiments? and (2) Why carry them out in space? The reason for asking the first question was the feeling that any proposed experiment should answer an important open question in atmospheric science and should be expected to yield significant results. In some cases, however, a proposed experiment may not answer pending questions, but will open up new areas of research not available to investigators in an earth gravity environment (an example of this is the study of liquid-liquid separation phenomena). The second question highlights the fact that we should only consider experiments which cannot be carried out under normal earth gravity. The members of the group were quite aware of the high costs and extraordinary difficulties associated with experiments in space. An underlying theme of the discussion was, "If it can be done on Earth, it should be done on Earth."

A number of experiments were described and discussed by the participants and are listed below. The experiments included in this report are not intended to be a comprehensive or final list of the important atmospheric particle experiments to be carried out in the Space Station. By the time a low gravity facility is available, other experiments unenvisaged by us may well have priority. Nevertheless, these experiments are representative of the types that atmospheric scientists will wish to perform, and it is reasonable to assume that the constraints on the experimental set-ups will not be considerably different from those discussed here. Consequently, the facilities required for these experiments will almost certainly be appropriate for future atmospheric experiments involving particles.

Of particular interest to atmospheric scientists is the formation of clouds, fogs, and aerosols. A low gravity facility is of particular value to cloud physicists because clouds and cloud particles can be studied without the effects of convection and particle settling. Individual particles and populations of particles can be studied for long periods of time. An especially important project is the analysis of very large drops. These cannot be generated in the presence of gravity due to spontaneous break-up. Clouds cannot be duplicated in the low gravity environment because their development depends on gravity and convection, however, one can isolate certain processes that are not controlled by gravity and study these processes over long periods of time. Similarly, processes occurring in atmospheric aerosols such as coagulation, growth, and chemical reactions, can be studied without strong gravitational influences and this will shed light on the basic physical processes which are taking place.

These experiments cannot be carried out on Earth because they require that no convection take place. Furthermore, these experiments require low gravity to produce long suspension times. The experiments will shed light on important areas of concern, including cloud growth, scavenging of atmospheric aerosols and an understanding of planetary cloud optical properties.

4.2 Suggested Experiments for Space Station

4.2.1 Growth of Liquid Water Drop Populations

Theory predicts that the size distribution of a population of liquid water droplets will become narrow due to continued condensation. Various processes, such as radiative cooling/heating, *etc.*, may affect the actual development of the size distribution. On Earth, the experimental determination of the time development of a droplet population is affected by sedimentation in which the larger droplets are continually being removed to a different region of space, not only changing the size distribution at a point (by removal of large particles) but also changing the size distribution by the coalescence of smaller particles with the sedimenting larger particles. Furthermore, the condensation process is accompanied by the liberation of latent heat. In a gravitational environment this generates convection which will also affect the droplet size distribution. The growth of a population of cloud droplets is of particular interest in understanding the formation of rain in warm clouds.

4.2.2 Coalescence (not collision)

Most experiments in 1 g measure collection which is the product of collision and coalescence efficiencies. However, most experiments cannot separate collision from coalescence. When only coalescence is measured in 1 g, the drops are normally suspended from a support which inhibits some of the natural deformations and oscillations. In addition, only relatively small drops (<2 mm radius) can be used in these experiments which makes it difficult to observe the deformations. A micro-gravity environment is a very suitable place in which to study the coalescence of any size drop. Using very large drops, it will be possible to study the drop deformation prior to contact and the mechanism by which first contact is made. By using different size drops, one may be able to extrapolate down to smaller drops such as those found in clouds.

4.2.3 Drop Breakup

The breakup of drops due to collision can be studied with large drops. This will permit a better evaluation of the hydrodynamic effect occurring during the merger and subsequent separation of the drops than is presently known. The size distribution of the breakup fragments can be studied as well. Such an experiment could be very important for the understanding of the development of rain drop spectra between cloud base and ground.

4.2.4 Breakup of Freezing Drops

It has been suggested that ice multiplication (or ice enhancement as it is sometimes called) results from the fragments of ice which are ejected from freezing drops. A microgravity environment is ideal for studying this phenomenon as the number of fragments ejected from the drops can be analyzed. The effect of the supersaturation around the freezing drop on the interstitial aerosols can be studied as a function of time.

4.2.5 Ice Nucleation for Large Aerosols or Bacteria

Large aerosols are difficult to suspend in cloud chambers. In microgravity it would be possible to determine whether large particles (even large ice nucleating bacteria) act as condensation-freezing or as deposition nuclei when exposed to a water saturated environment.

4.2.6 Scavenging of Gases (e.g., SO_2 oxidation)

By floating large drops for long periods of time, SO_2 absorption can be studied. Similar experiments with smaller drops (smaller than 50 μm) could very well duplicate the occurrence in 1 g, since the fall speed of small drops is small. Even though some convection mixing occurs, the diffusion of the reactants to the drops is sufficient to keep up with the relatively slow oxidation rate.

4.2.7 Phoretic Forces: Thermophoresis Versus Diffusiophoresis

In an environment where a temperature gradient is present, aerosol particles will experience thermophoretic forces due to the difference in the momentum transferred by gas molecules impacting them from both the warm and cold sides. Therefore, aerosols will move down the temperature gradient, toward the colder region. A diffusiophoretic force is the one given to an aerosol particle due to concentration gradients in the gaseous mixture. During condensation, the diffusiophoretic forces will cause aerosol particles to move towards the condensing surface. On the other hand, the release of latent heat will raise the surface temperature driving the aerosol away from it due to the thermophoretic forces. Some experiments show that thermophoretic forces dominate. Therefore, when ice and water co-exist and ice grows at the expense of the drops, it is expected that interstitial aerosols will move to the evaporating drops. However, some experiments report a dust free region around evaporating drops, suggesting that diffusiophoresis dominates. Similarly, when ice crystals, suspended from a fine fiber, are allowed to flow, aerosol particles are collected on their surfaces. The lack of agreement among experimenters may be a direct result of the heat conducted away through the fiber supporting the drop or the ice crystal. The conduction of heat away from the surface reduce the temperature gradient and hence reduce the thermophoretic force, making the diffusiophoretic force dominant.

An experiment in microgravity conditions could resolve this question by having ice crystals, water drops and aerosol particles all floating in a water saturation environment. The concentration gradients and velocities of the aerosol particles could be measured with the aid of a Doppler laser or by other means. It is important to note that these phoretic forces are useful in the scavenging of the particles that are too large to be affected by diffusion and are too small to be captured by gravitational impaction.

4.2.8 Rayleigh Bursting of Drops

This process is thought to be the trigger to lightning. Here drops are charged to the limit at which they burst. This limit is achieved when the electrical stresses just equal the surface tension forces. There is some evidence to suggest that drops larger than 7 mm diameter will disrupt when their surface tension energy is exceeded by their electrical energy. This will also happen to very small droplets. For drops in the size range from 50 μm to 7 mm, corona discharge precedes breakup. In 1 g large drops cannot be studied since they cannot be produced, while small ones disrupt too easily. With microgravity we have the opportunity to study large drops and their disruption.

4.2.9 Charge Separation Due to Collisions of Rimed and Unrimed Ice

In this experiment a large ice crystal, previously grown by rime, would be introduced into a cold chamber and floated. Small diffusionally grown ice crystals would be forced to impact the floating one. Once rebounded, all crystals could be captured in a Faraday cage for analysis of the charge that has been separated. The microgravity condition makes it possible to float nonspherical crystals and analyze them after impact with other crystals. Since this kind of charging is thought to be the main mechanism of charge separation in thunderclouds it is important to test it with different crystal sizes and under different temperatures.

4.2.10 Coalescence

Coalescence is the process by which particles with differing velocities impact one other and unite to form a larger particle. This process is essential to the formation of rainfall and lightning as well as to the removal of particles in dense clouds such as those from larger fires or volcanic eruptions. Unfortunately, these interactions are so complex that no theory exists which is adequate even for spheres of all sizes. Indeed, various theories strongly disagree with one other and with data. The experimental data base is also strongly restricted.

When particles of moderate size approach one other, their coalescence may be inhibited by hydrodynamic interactions. For example, a small particle may simply follow the flow field around a larger one and never make physical contact so that coalescence fails to occur.

Unfortunately, experiments to study coalescence are very difficult on earth. The difficulty is partly due to the inability to easily observe particles falling in the earth's gravity field because they move across the sampling chamber so rapidly. Suspending the particles in the gravity field is unsatisfactory too, because it

leads to changes in the flow fields. The experimental difficulty is also partly due to the fact that turbulent coalescence cannot be easily separated from gravitational coalescence because one cannot eliminate the gravitational interaction. Also, one cannot examine a large range of collision velocities because on earth, the velocities are fixed by gravity. All of these problems might be eliminated by performing studies in a microgravity environment.

We envision studying turbulent coalescence, coalescence due to differential velocities, and Brownian coalescence and the interactions between each each of these processes as functions of particle velocity, density, and particle shape. The major advantages of doing this in space is that the particle differential velocity can be controlled, and the particles can be easily contained while the experiment is being conducted.

4.2.11 Charged Drop Dynamics

In charged drop dynamics we are interested in vibrational and rotational dynamics and the stability characteristics of the drop. Furthermore, we wish to explore Rayleigh bursting, corona discharge and other such phenomena at all levels of charges on the drop.

The electrohydrodynamical problems have been computer simulated primarily by Scriven, Brown and coworkers. The results of these studies are waiting for experimental verification.

In order to carry out charged drop dynamics experiments it is necessary to position a large drop at a desirable position, and its charge levels and surface tension should be accurately measured or monitored. Any levitation force in 1 g (either electrostatic or acoustic) creates large perturbations to the system which in turn creates great theoretical difficulties in interpreting the results.

Knowing the fundamental dynamic characteristics of a charged liquid drop will lead to a clear understanding of electrohydrodynamical problems in cloud physics, shell technology (core centering effect, *etc.*), aerosol physics and the scavenging of aerosol.

4.2.12 Growth of Particles in Other Planetary Atmospheres

In addition to usefulness of a microgravity environment for studying the properties of stratospheric (H_2SO_4) and tropospheric (water) clouds on the Earth, an even wider range of questions can be addressed with such a facility regarding the clouds that occur on other planets. Recent space missions have revealed the presence of bright zones, and darker belts of troposheric clouds on Jupiter and Saturn. Stratospheric aerosols have also been observed on these planets and on Titan. Equilibrium chemical models suggest that the uppermost layer of tropospheric clouds on Jupiter and Saturn consist of NH_3 ice crystals, but this suggestion remains to be confirmed by spectroscopic observations. Even if ammonia is a major constituent of these clouds, it cannot be the only constituent as pure ammonia ice clouds would be white at visible wavelengths in contrast to the color observed on these planets. Similarly, several constituents have been suggested for the photochemically produced stratospheric aerosols on these planets including polyacetylenes, N_2H_4, and P_2H_4, but a definitive identification remains to be made. For these planets even the basic composition of the cloud and aerosol particles are still uncertain. A host of other questions concerning the vertical and horizontal distribution, shapes, crystal types, production and transport remain to be answered.

A considerable amount of indirect information that bears on these questions has been collected in measurements of the photometry and polarimetry of the sunlight reflected from the Pioneer and Voyager missions, and more such data are anticipated from the Galileo orbiter and probe. Scattering calculations have been used to convert the observations of multiply-scattered light to constraints on the single-scattered phase function and polarizing properties of these cloud particles. However, because the cloud particles exist as solid crystals rather than as spherical liquid droplets at the low temperatures on the outer planets, Mie scattering calculations cannot be used to convert their single-scattering optical properties to constraints on their size distributions and refractive index. Rather, a systematic program of scattering measurements of candidate cloud particles is needed for comparison with the existing observations.

Some preliminary measurements of the crystal habits of ammonia ice as functions of temperature and pressure are just now beginning to be made. These measurements must:

1. Map the crystal habits formed by ammonia and other candidate materials as functions of temperature and degree of saturation.

2. Simultaneously measure the optical scattering properties of these particles (intensity and degree of polarization) as functions of scattering angle (preferably at more than one wavelength).

3. Provide a high-resolution record (photographs) of the crystal shape and size distribution whose optical properties have been measured.

4. Extend over a range of temperature appropriate for Jupiter and Saturn (down to 80 K).

The program of measurements currently under way has produced some important results for some range of conditions, but also suffers from fundamental limitations. A real cloud of large particles (including a range of particle sizes and orientations) is difficult or impossible to measure because the particles fall out before they grow to large sizes. Measurements in a low-g environment would eliminate this fundamental difficulty.

Further, because the production rates for producing photochemical smog particles tend to be lower than setting rates, this material can accumulate in the bottom of the chamber instead of producing a cloud of aerosols whose scattering properties can be measured. Existing studies have been limited to calculations for spherical shapes using the properties of samples from the bulk material scraped from the bottom of the reaction chamber. Production in a low-g environment would permit scattering measurements to be made of aerosol particles as aerosols before they settle on the chamber walls.

4.2.13 Freezing and Liquid-Liquid Evaporation

Unless efforts are made to ensure equilibrium, phase changes (*e.g.*, condensation, boiling, crystallization) involve the nucleation of the new phase. All nucleation processes have a random component. Thus, if one cools a particular drop it will subcool to slightly different extents each time. However, a large population of drops will have the same freezing temperature distribution each time one subcools it. For this reason one tries to devise experimental approaches that study large populations. Unfortunately, this is not always possible.

One example of a phase transition that is difficult to study on Earth is the effect of charge on the separation of a liquid mixture into two separate liquids as cooling occurs (*i.e.*, liquid-liquid nucleation). The two liquids always have very different densities. As a result, as soon as the phase separation occurs, the liquids separate. One needs to then mechanically re-mix the system since diffusion velocities in liquids are very small. This method imposes enormous additional difficulties. In addition, it is not possible to study the growth and aggregation of the charged particles. Microgravity will greatly reduce the difficulties since the rate of fall of the dense component will be much smaller. One can thus detect the nucleation by light scattering, then reheat the liquid and allow diffusive mixing to take place.

4.3 Required Capabilities of an Orbital Facility

To carry out these experiments which are important to the study of atmospheric science will probably require more than one experimental chamber or an adaptable chamber. For example, for light scattering (*i.e.*, optical properties) of aerosols, it is most convenient to have a chamber with many windows, or even better, a continuous window running all the way around the circumference of the chamber. On the other hand, to maintain a saturated environment within the chamber, the window area should be reduced as much as possible.

The chamber need not be too large. Although wall effects may be an important constraint, particle stabilization can probably be effected by electric or acoustic techniques. Consequently, a chamber of the order of 250 cm^2 by $\sim$50 cm in depth would probably be sufficiently large for most experiments.

Provisions should be made for optical sensor and photographic equipment to be integrated with the experimental chamber. In addition, the chamber should allow for a controlled humidity and temperature (from about 80 K to 300 K). There should also be the possibility of carrying out high temperature experiments. For heterogeneous chemistry experiments, appropriate chemical apparatus will be required. The mechanical removal and insertion of particles should be possible, and the chamber should be easily connected to an aerosol generator.

Requirements of these types of studies on a microgravity particle measuring facility would include:

1. Provision to cool a chamber to temperatures as low as 80 K and maintain it at specified low temperature gradients.
2. The presence of a double cylindrical window in the chamber to permit scattering measurements over a range of scattering angles (10° to 165°).
3. Provision to record the size and shape of crystals in the chamber to as small a size as possible (1 m).
4. Light sources at several wavelengths (at least red and blue) to illuminate the crystals, a system to control the polarisation state of the incident beam and a set of detectors spaced at angles to make the scattering measurements.
5. A gas handling system for admitting controlled amounts and mixtures of gas.
6. Windows and UV lamps for producing photochemical aerosols.

Probably such a low-temperature chamber would be distinct from a high temperature facility which could be used to study condensation of refractory materials. It is also possible that three distinct types of chambers are desirable to allow for three different types of experiments:

1. High temperature refractory condensation ($T > 1000$ K).
2. Water liquid and ice cloud physics studies ($-50°C < T < 20°C$) possibly without any scattering measurements.
3. Optical scattering measurements ($80 \text{ K} < T < 290 \text{ K}$).